EXPLORING THE POTENTIAL OF NANOFILLERS FOR ADVANCED THIN FILM NANOCOMPOSITE FORWARD OSMOSIS MEMBRANES FABRICATION

MOHAMMAD GHANBARI

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Petroleum Engineering)

Faculty of Petroleum & Renewable Energy Engineering
Universiti Teknologi Malaysia

OCTOBER 2015

DEDICATION

I dedicate this dissertation to my beloved family;

my dear father;

and my merciful mother, for her encouragement

ACKNOWLEDGEMENT

First of all, I would like to thank almighty Allah for establishing me with strength and faith, and giving me the sight to realize myself. Incontrovertibly, I owe my supervisor, Professor Dr. Ahmad Fauzi Ismail a great deal of debt, for his kindness and guidance throughout my entire research. He encouraged me by his constructive advices and intellectual supports during my doctoral period. His friendly personality has always created a positive atmosphere and motivated me to work. My sincere appreciation also extends to my co-supervisor Dr. Lau Woei Jye who has been the most energetic and great inspiration to me in my research and gave me the inspiration to keep on the right direction during my research. Without him, I could never accomplish my study smoothly. It is with immense gratitude that I acknowledge Professor Dr. Takeshi Matsuura for his fundamental and invaluable direction, guidance and assistance. I have learned from him not only how to perform and interpret experiments but also how to think and move the project forward. His exceptional insights into engineering have immensely helped me to enrich my knowledge. In addition, I want to extend my thanks to the all Advanced Membrane Technology Research Centre (AMTEC) members for their friendship, invaluable assistance and giving me invaluable advice during throughout this period.

I would also like to thank all those who were involved directly or indirectly in the completion of this project. My thanks also go to my parents, for their blessings and encouragements throughout my life. Finally yet importantly, I would like to express utmost appreciation to my lovely and kind siblings for their love, support and encouragements throughout my life.

ABSTRACT

Novel and promising forward osmosis (FO) is a membrane-based separation with significant potentials for the desalination process. While this technology offers various benefits, overcoming its internal concentration polarization (ICP) and membrane fouling in polyamide (PA) skin layer remain as a challenge. In this study, three types of novel thin film nanocomposite (TFN) membranes were synthesized by either coating a typical PA film over the surface of substrate made of polysulfone–halloysite nanotubes (HNTs) or embedding HNTs and titanium dioxide (TiO_2)/HNTs nanocomposites into PA thin layer formed over a typical polysulfone (PSF) substrate. These approaches aim to reduce membrane fouling and/or ICP during FO applications. In the first stage of this study, both hydrophilicity and porosity of the substrate were increased using HNTs. The results obtained from filtration experiments showed that the TFN membrane prepared with incorporation of 0.5 wt% HNTs (TFN 0.5) demonstrated the most satisfactory results by exhibiting high water permeability and low reverse solute flux in both FO and pressure retarded osmosis (PRO) configurations. This improvement can be ascribed to the fact that the structural parameter (S value) of TFN membrane is much lower compared to that of control thin film composite (TFC) membrane (0.37 vs 0.95 mm), leading to reduced ICP effect. In the second stage of this study, both hydrophilicity and surface roughness of TFN membranes increased with incorporation of HNTs into PA layer. In the FO mode, the fabricated TFN FO membrane in this study exhibited significantly higher fouling resistance compared to the control TFC membrane. As an indication to reversibility of fouling in TFN FO membrane, it was also found that more than 96% permeate flux could be recovered after a simple water rinsing process. In the third stage of this study, TiO_2/HNTs nanocomposites synthesized via one-step solvothermal method were used as nanofillers in the preparation of TFN membranes for the FO application. With respect to separation performance, it was discovered that the TFN membrane incorporated with 0.05% (w/v) TiO_2/HNTs (TFN 0.05) exhibited the best performance due to its high water permeability and low reverse solute flux when tested using 10 mM sodium chloride (NaCl) feed solution and 2.0 M NaCl draw solution under two different membrane configurations. Compared to the control membrane (without TiO_2/HNTs incorporation), the fabricated TFN 0.05 membrane could offer up to 90% higher water flux and exhibited significantly better antifouling affinity against bovine serum albumin (BSA). The results revealed that fouling in the TFN 0.05 membrane was completely reversible. As a conclusion, it was found that modifying the PA skin layer of composite membrane using TiO_2/HNTs as nanofillers could give the most promising results, improving not only membrane permeability and selectivity but also its anti-fouling property.

ABSTRAK

Proses osmosis hadapan (FO) yang novel adalah satu teknik pemisahan berasaskan membran yang berpotensi besar untuk proses penyahgaraman. Walaupun teknologi ini menawarkan pelbagai kelebihan, cabaran utama yang perlu diatasi adalah polarisasi kepekatan dalaman (ICP) dan kotoran membran pada lapisan aktif poliamida (PA). Dalam kajian ini, tiga jenis novel membran filem nanokomposit nipis (TFN) telah disintesis sama ada melalui kaedah penyalutan filem PA di atas permukaan substrat yang diperbuat daripada polisulfona- tiub nano haloisit (HNTs) atau menggabungkan HNTs dan titanium dioksida (TiO_2)/HNTs nanokomposit dengan lapisan nipis PA yang terbentuk di atas substrat polisulfona (PSF). Pendekatan ini bertujuan untuk mengurangkan kotoran membran dan/atau ICP semasa proses FO. Pada peringkat pertama kajian, kehidrofilikan dan keliangan substrat PSF telah meningkat selepas penambahan HNTs. Keputusan yang diperoleh daripada kajian turasan telah mendapati membran TFN yang diperbuat daripada 0.5% berat HNTs dalam substrat (TFN 0.5) menunjukkan fluks air yang tinggi dan fluks bahan terlarut yang rendah dalam konfigurasi FO dan konfigurasi tekanan osmosis terbantut (PRO). Peningkatan ini disebabkan oleh parameter struktur (nilai S) untuk membran TFN yang jauh lebih rendah berbanding dengan membran kawalan filem komposit nipis (TFC) (0.37 vs 0.95 mm), yang mengakibatkan kepada pengurangan kesan ICP. Pada peringkat kedua kajian, kehidrofilikan dan kekasaran permukaan membran TFN meningkat dengan penambahan HNTs ke dalam lapisan PA. Pada mod FO, membran TFN FO mempunyai rintangan kotoran yang lebih tinggi berbanding dengan membran kawalan TFC. Bagi membuktikan keboleh-balikan kotoran dalam membran TFN FO, hasil kajian menunjukkan bahawa lebih daripada 96% fluks boleh diperoleh semula selepas proses pembilasan air yang mudah. Pada peringkat ketiga kajian, nanokomposit TiO_2/HNTs yang disintesis melalui kaedah solvoterma telah digunakan sebagai pengisi-nano dalam penyediaan membran TFN untuk proses FO. Hasil kajian menunjukkan bahawa membran TFN yang digabungkan dengan 0.05% (berat/isipadu) TiO_2/HNTs (TFN 0.05) mempunyai prestasi yang terbaik dengan kebolehtelapan air yang tinggi dan fluks bahan larut balikan yang rendah apabila diuji menggunakan 10 mM natrium klorida (NaCl) larutan suapan dan 2.0 M NaCl larutan luaran pada dua konfigurasi membran yang berbeza. Berbanding dengan membran kawalan (tanpa TiO_2/HNTs), membran TFN 0.05 mampu menghasilkan fluks air 90% lebih tinggi dan sifat anti-kotoran terhadap serum bovin albumin (BSA) yang jauh lebih baik. Hasil kajian juga menunjukkan bahawa kotoran pada membran TFN 0.05 boleh berbalik. Kesimpulannya, pengubahsuaian lapisan aktif PA membran komposit menggunakan TiO_2/HNTs sebagai pengisinano boleh meningkatkan bukan sahaja kebolehtelapan dan kememilihan membran tetapi juga sifat anti-kotorannya.